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Published in:
Proceedings of the International Conference on Radioecology and Environmental Radioactivity

Publication date:
2014

Document Version
Peer reviewed version

Improvement of radiological consequence estimation methodologies for NPP accidents in the ARGOS and RODOS decision support systems through consideration of contaminant physico-chemical forms

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INTRODUCTION / BACKGROUND

The European standard computerized decision support systems RODOS and ARGOS, which are integrated in the operational nuclear emergency preparedness in practically all European countries, as well as in a range of non-European countries, are highly valuable tools for radiological consequence estimation, e.g., in connection with planning and exercising as well as in justification and optimization of intervention strategies. Differences between the Chernobyl and Fukushima accident atmospheric release source terms have demonstrated that differences in release conditions and processes may lead to very different degrees of volatilization of some radionuclides. For instance, the Chernobyl accident released a lot of radioruthenium, which reached faraway areas predominantly in the form of slightly submicroneous particles with a low deposition velocity (Reineking et al., 1987). However, a fraction of the ruthenium aerosol reaching for instance Germany and Norway was of a distinctly different size (reaching AMAD of the order of 10 μm), indicating a skewed size distribution of the released Ruparticles from the fuel. White inclusions containing Ru were also identified imbedded in U fuel particles. In connection with the Fukushima release, no radioruthenium was observed. It should here be noted that the volatility of Ru at certain temperatures strongly depends on the oxidation of the source material (Kashparov et al., 1996; Hunt et al., 1994). As the level of contamination (and longer term dose) in an area is directly proportional to the contaminant deposition velocity, and the deposition velocity strongly depends on aerosol size distribution, it is crucial for the parametric source term input to the European decision support systems to adequately reflect physico-chemical characteristics specific to the particular release type in question. Physico-chemical forms of contaminants released to the atmosphere can also greatly affect the post-deposition environmental migration of the contamination. An example from the Chernobyl accident of the significance of this is that uranium fuel particles (U-Zr, reduced U) released during the explosion had properties significantly different from those released during the subsequent fire (oxidised U), of importance for the ecosystem transfer. . This is reflected in the very different particle weathering rates and the subsequent remobilization of associated radionuclides. The initial matrix characteristics of the contaminants, as well as environmental parameters like pH, determine for instance the particle weathering time functions, and thus the environmental mobility and potential for uptake in living organisms. As ICRP recommends optimization of intervention according to residual dose, it is crucial to estimate long term dose contributions adequately.

RESULTS / DISCUSSION

In the EURATOM FP7 project PREPARE, an effort is made to integrate information on physico-chemical forms of contaminants in scenario-specific source term determination, thereby enabling consideration of influences on atmospheric dispersion/deposition, post-deposition migration, and effectiveness of countermeasure implementation. A first step in this context was to investigate, based on available experience, the important physico-chemical properties of particles containing radiocontaminants that might potentially be released to the atmosphere in different conceivable nuclear power plant accidents. Reactor experts have evaluated each of the 46 different conceivable release types and associated release source terms (radionuclides) currently considered relevant for existing reactors according to the RODOS decision support system, and considered the release conditions including potential for oxidation and peak temperatures. The review concluded that all considered scenarios would involve oxidizing conditions, and that the type of powerful explosion that occurred in Chernobyl and led to dispersion of non-oxidised contaminants over a large land area would not be relevant with present day nuclear power plant designs.

An important source of information to the project is constituted by the results of the PHEBUS FPT0/1/2/3 NPP accident test programme, which was performed by the French institute IRSN in collaboration with a range of other organisations from Europe, USA, Canada, Japan and Korea. This work, which was done over a number of years since 1988 (see, e.g., Gregoire et al., 2008), was carried out to investigate key severe accidents and associated releases that might occur at light water cooled reactors typical of those in operation in member countries of the European Union. Of particular interest in the present context is a table with information on readily water soluble fractions of radionuclides of different important elements that could be released to the atmosphere. For instance a very large fraction of the caesium aerosols released to the atmosphere (~95 %) was generally found to be readily soluble in water. This is what would be expected in connection with releases of small submicron particles formed through evaporation followed by condensation/nucleation, as was the case in connection with oxidized U in the Chernobyl and Fukushima releases, as well as with American blast experiments involving caesium in liquid form (Andersson et al., 2013). The general similarities in release processes for caesium were underlined by results of impactor measurements of the aerosol size of radiocaesium contaminants at Tsukuba, Japan (Kaneyasu et al., 2012), which agree with what was recorded by, e.g., Reineking et al. (1987) after the Chernobyl accident.

It is, however, also clear from the PHEBUS test material that contaminant aerosols of other important elements, including strontium and cerium, which were in the Chernobyl release generally found to be considerably less volatile, will generally be much less readily soluble in the environment. Kashparov et al. (1996, 2004) thoroughly investigated the time dependence of the fraction of radiostrontium dissolved from both oxidized and non-oxidised fuel particles in soil areas contaminated by the Chernobyl accident. They found that the strontium mobilization after deposition of both oxidized and non-oxidised fuel particles in the environment was a slow process. This weathering rate (in units of y^{-1}) was for the oxidized particles approximated by the expression $k = 23*(10^{(-0.35*pH)}+1.1666667*10^{(-4.55)}*10^{(0.3*pH)})$. Presumably, similar weathering rates apply for other radionuclides from similar low solubility matrix. This implies for example that at pH 6, only about half of the radiostrontium will have been leached from oxidized fuel particles after a period of ten years. This is in sharp contrast to the current modelling in the food dose and external dose models currently applied in RODOS and ARGOS, where all contaminants are assumed to be readily soluble from the moment of deposition in the environment. This means that for instance contaminant crop uptake over the first few years can be greatly overestimated by the decision support systems, whereas long term doses (including residual doses following implementation of countermeasures) would be as greatly underestimated. This problem is being addressed by revising all contaminant migration parameters in the decision support systems and making them considerably more release scenario specific. A useful overview of information on contaminants released in the past in different types of major nuclear and radiological releases has also been worked out in the PREPARE project, with focus on the types of events, temperatures and pressures during releases, particle matrix and density, contaminant weathering rates / solubilities, particle sizes released, etc.

It should be noted that countermeasure efficiency is, both in the computerized decision support systems and in the European handbooks for assisting in recovery of contaminated areas, to a very great extent based on Chernobyl experience for cationic caesium, which is known to be retained strongly and selectively on many types of environmental surfaces. An effort has been made in the PREPARE project to examine the efficiency of countermeasures in relation to contaminants with different physico-chemical characteristics. For instance, although different mechanisms can bind contaminants to surfaces, Sartor et al. (1974) found experimentally that removal of contaminants in particle form from impermeable road surfaces was largely independent of particle size when the particles were larger than about 10 µm. Smaller particles are however generally increasingly difficult to remove as they can enter cavities in structures, and be much less prone to weathering. For example a simple countermeasure such as firehosing on street surfaces would only be expected to reduce the contamination level for 0.7 µm particles by about a factor of 2, but for particles in the size range of 4 µm, the contamination level could be reduced by as much as a factor of 6. Such new contaminant-specific countermeasure effectiveness factors are being defined for use in RODOS and ARGOS.

CONCLUSIONS

The reliability of dose prognoses made with the European decision support systems for addressing the consequences of nuclear or radiological releases to the atmosphere is being improved. The physico-chemical forms of each contaminant is being taken into account for the given type of release scenario, in the choice of parameter values for environmental mobility models for contaminants in the atmosphere as well as after deposition in inhabited and agricultural areas. Also decontamination efficiency factors are revised.

This work was partly supported by the CEC: EURATOM FP7 project PREPARE (Grant agreement no: 323287) and by Research Council of Norway through its Centres of Excellence funding scheme, project no. 223268/F50.

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